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THE PROBLEM OF UTILIZING SEMICONDUCTING LIQUIDS FOR IMPREGNATING PAPER CONDENSERS

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[Figures are appended.]

Experiments were started in 1941 by Docent A. M. Lazarev at the Leningrad Polytechnical Institute but were not completed due to the war, and his subsequent death. In 1947, Clark (see Electricity, No 10, 1947, p 77 of the United States published an article describing similar experiments. He stated that the loss angle in impregnated paper increases sharply at first, as the conductivity of the impregnating substance increases; i.e., when its specific resistance is lowered, and it attains its maximum when the specific resistance reaches 10^9 ohm/cm, and then falls. When the specific resistance is 10^8 ohm/cm, i.e., sharp decrease in comparison with the specific resistance of ordinary dielectrics, the loss angle passes through its minimum, after which it rises again with increasing conductivity of the impregnating substance. Depending on the type of semiconducting liquid and the type of paper, the minimum value for the power factor of the paper which is impregnated with this liquid is approximately 3 to 6 percent. This makes it possible to utilize such a dielectric in low-voltage condensers (110-220v) at standard frequencies.

The specific volume of condensers impregnated with semiliquid impregnators is about one third that of paper-oil condensers for similar voltages.

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The complex nature of the relationship cos $\varphi = f(\rho)$ which was obtained by Clark shows up individual anomalies (according to Clark) in the behavior of cellulose insulation that is impregnated with semiconducting liquid. Clark makes no attempt to investigate the physical aspects of the phenomenon which could explain the character of the relationship obtained and the presence of maximum and minimum values in the experimental curves for cos φ.

In addition, it can be stated that the particular nature of the cos $\varphi = f(\rho)$ relationship, which characterizes the occurrence of maximum and minimum values of cos φ when the value of specific resistance ρ is decreased from 10^{12} to 10^{6} ohm/cm, is completely regular and does not in any way reveal unknown anomalies in the performance of impregnated paper.

In examining the relationship between the dielectric constant of impregnated paper and the dielectric constant of the impregnating substance, we were able to determine that paper condensers impregnated by liquids can be compared to a system consisting of two condensers connected in series: C_m is the theoretical capacitance based on polarization of the impregnating substance, and C_k is the theoretical capacitance based on the polarization of the cellulose (see Electricity, No 1, 1949). With impregnation by a semiconducting substance, it is necessary to include in this circuit an effective resistance R parallel to the capacitance C_m (Figure 1). This resistance represents the conductivity of the semiconducting substance. The conductivity of the cellulose can be disregarded in the first approximation.

The following expressions are obtained from the symbolic method of the circuit in Figure 1:

$$Z_\Sigma = Z_M + Z_K = \frac{R - jR^2\omega C_M}{1+R^2\omega^2 C_M^2} - j\frac{1}{\omega C_K} =$$

$$= \frac{R}{1+R^2\omega^2 C_M^2} - j\,\frac{1+R^2\omega^2 C_M(C_K+C_M)}{\omega C_K(1+R^2\omega^2 C_M^2)} = r_\Sigma - j\frac{1}{\omega C_\Sigma}.$$

The tangent of the loss angle for this equivalent circuit will equal:

$$\tan\delta_\Sigma = \omega r_\Sigma C_\Sigma = \frac{\omega R C_K}{1+R^2\omega^2 C_M(C_K+C_M)} \qquad (1)$$

The above expression (1) shows that tan δ_Σ reduces to 0 for $R = 0$ or $R = \infty$. This proves that for certain conditions of the specific resistance of the substance, the angle of the impregnated paper must pass through some maximum value. In the case of Figure 1, the overall losses approach zero for large values of specific resistance ρ due to the fact that they are very small in condenser C_m and are negligible in condenser C_k. For small values of specific resistance ρ, condenser C_m with the large losses is short-circuited, leaving only condenser C_k which has no losses; i.e., the over-all losses of the system again approach zero.

Let us assume that the dielectric constants of the cellulose and of the impregnating substance are exactly equal, $\varepsilon_k = \varepsilon_m = 7$; the thickness of the paper is $d = 10$ microns $= 10^{-3}$ cm (similar to values used by Clark in his experiments); and the ratio of the volume of the impregnating substance to the volume of the paper is $a = 0.35$ (paper of normal density).

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The following formulas are obtained when making calculations for one sq/cm of paper surface:

$$C_K = \frac{\varepsilon_K}{4\pi(1-a)d} \cdot 1.11 \cdot 10^{-12} = 0.96 \cdot 10^{-9}\ F/cm^2;$$

$$C_M = \frac{\varepsilon_M}{4\pi a d} \cdot 1.11 \cdot 10^{-12} = 1.77 \cdot 10^{-9}\ F/cm^2;$$

$$R = \rho a d = 3.5 \cdot 10^{-4} \rho,$$

where ρ is the specific resistance of the semiconducting substance in ohm/cm.

If we insert numerical values in formula (1) the following expression is obtained

$$\tan \delta_{\Sigma} = \frac{1.055 \cdot 10^{-10}\rho}{1 + 5.82 \cdot 10^{-21}\rho^2} \qquad (1')$$

Results of the calculations from formula (1') are shown graphically in Figure 2 by curve A.

The graph differs from the experimental data not only in that it has no minimum but also in that the maximum corresponds to $\rho \approx 10^{10}$ ohm/cm and not to $\rho \approx 10^{9}$ ohm cm. Moreover, the absolute value of the loss angle at the maximum is very low in comparison with experimental data ($\tan \delta = 0.635$ which corresponds to the value $\cos \varphi = 0.537$, while Clark obtained $\cos \varphi = 0.870$). This difference can be explained by the fact that the circuit in Figure 1 does not represent exactly the true nature of the phenomenon.

In the circuit the cellulose is represented in the form of a compact layer which separates the impregnating substance from one of the faces. Actually, however, the condenser paper contains open pores of minute cross sections; their presence is established by the fact that air can pass through the paper. During the impregnation process, the impregnating substance fills these pores, but this factor can be ignored when computing the dielectric constant of the impregnated paper because of the fact that the volume of impregnating mass in these pores is very minute and has no noticeable effect on the over-all polarization of the system under study. But it must be kept in mind that if the paper is impregnated with a liquid which has a relatively low specific resistance, the resistance of the pores filled with the impregnating substance may be sufficiently small to cause a noticeable change in the over-all loss angle, notwithstanding the small cross section of these pores. It is therefore necessary to use a circuit which is more complicated than that represented in Figure 1. In Figure 3 the capacitance C_K is shunted through several resistances ηR. The value of η must be several times greater than one, due to the fact that the resistance of the substance in the pores is greatly increased because of their small cross section:

$$\eta = \frac{\rho \frac{d_K}{S_K}}{\rho \frac{d_M}{1}} = \frac{1-a}{a} \frac{1}{S_K},$$

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where S_k is the sum total of the cross sections of the open pores per sq cm of paper.

The following formula is obtained for Figure 3:

$$Z_\Sigma = Z_M + Z_K = \frac{R - jR^2\omega C_M}{1 + R^2\omega^2 C_M^2} + \frac{\eta R - j\eta^2 R^2\omega^2 C_K}{1 + \eta^2 R^2\omega^2 C_M^2} =$$

$$= \frac{R[1+\eta+\eta R^2\omega^2(C_M^2+\eta C_K^2)]}{(1+R^2\omega^2 C_M^2)(1+\eta^2 R^2\omega^2 C_K^2)} - j\frac{\omega^2 R^2\{C_M+\eta^2 C_K[1+R^2\omega^2 C_M(C_K+C_M)]\}}{(1+R^2\omega^2 C_M^2)(1+\eta^2 R^2\omega^2 C_K^2)}$$

The tangent of the loss angle in this equivalent scheme will be equal to

$$\tan\delta_\Sigma = \frac{1+\eta+\eta R^2\omega^2(C_M^2+\eta C_K^2)}{\omega R\{C_M+\eta^2 C_K[1+R^2\omega^2 C_M(C_M+C_K)]\}}. \qquad (2)$$

An analysis of formula (2) shows that when $R = 0$, the value of $\tan\delta_\Sigma$ does not become zero but becomes infinite; i.e., the loss angle increases when there is a sharp decrease in the value of specific resistance ρ. This fact agrees with experimental data.

The following expression is obtained if we insert known values in formula (2), omitting the terms known to be negligible:

$$\tan\delta_\Sigma = \frac{1+\eta+1.11\cdot 10^{-20}\eta^2\rho^2}{\eta^2\rho(1.055\cdot 10^{-10}+6.17\cdot 10^{-31}\rho^2)} \qquad (2')$$

The coefficient η, which is used in expression (2'), is not known but we may assign certain values to it, i.e., to select certain small values S_k for the cross-section of the pores in the paper, and draw a set of curves for $\tan\delta_\Sigma = f(\rho)$ corresponding to various values of η and select that particular curve which best agrees with the experimental data. Two such curves, B and C, are shown in Figure 2. Curve B which is obtained for $\eta = 4{,}000$ ($S_k = 4.65 \cdot 10^{-3}$ sq cm per sq cm of paper) represents the minimum loss angle, which equals the lowest limit obtained by Clark in his experiments ($\cos\varphi \approx 0.03$). Nevertheless, in this case the minimum is obtained when the specific resistance ρ has a value of 10^8 ohm/cm instead of 10^7 ohm/cm. Curve C is obtained when $\eta = 18{,}600$ ($S_k = 1.10^{-4}$ sq cm per sq cm of paper.)

This curve shows that by increasing η we may displace the minimum toward the lesser values for specific resistance ρ, but in this case the loss angle has a value lower than that obtained in experiments, when it passes through its minimum. The right sides of curves B and C coincide with curve A; therefore, passing over to the circuit in Figure 3 did not give us an approximation to the experimental data in the part near the absolute value of the maximum loss angle and for values of ρ near the observed maximum. In addition, the use of this circuit permits us to uncover the rule controlling the appearance of the minimum in the curve describing the relationship: $\tan\delta_\Sigma = (\rho)$, although on the basis of computations, the point of the minimum will correspond to a higher value of specific resistance ρ in comparison with experimental data. To obtain calculated values which are almost equal to experimental values in this particular experiment, as also in several others, it can be assumed that Clark utilized in his measurements a strip of thin gauze in order to eliminate the possibility of short circuits of the electrodes due to the paper's conducting action -- which could easily occur in experiments using one layer of paper. Although, referring to the graph of cos φ = f (ρ), Clark gives no indication that he made use of a layer of gauze, it is implied that he conducted his experiments using a single layer of paper.

The presence of a gauze layer on the top surface of the paper greatly increases the thickness of the layer of impregnating substance. This makes it necessary to introduce a series of new constants for the formula based on Figure 3.

If we were to assume that the thickness of the gauze layer is 100 microns, then we derive the following expressions:

$$C_M = 5.98 \cdot 10^{-11} F/cm^2;\quad C_K = 0.96 \cdot 10^{-9} F/cm^2;$$

$$R = 1.035 \cdot 10^{-2} \rho \text{ and } \eta = 6.3 \cdot 10^{-2} \frac{1}{S_K}.$$

Substituting these new values in formula (2) we obtain the following expression:

$$\tan \delta_{\Sigma} = \frac{1 + \eta + 9.7 \cdot 10^{-18} \eta^2 \rho^2}{\eta^2 \rho (3.13 \cdot 10^{-9} + 2.01 \cdot 10^{-27} \rho^2)}. \qquad (2'')$$

Results obtained by calculations with formula (2") were satisfactory and lead one to believe that the supposition that a gauze layer was used is correct. Figure 2 shows the curves obtained by calculating with formula (2") for two values of η. Curve D was obtained for η = 100 ($S_k = 6.3 \cdot 10^{-4}$ sq cm per sq cm of paper) while for curve E, η = 500 ($S_k = 1.26 \cdot 10^{-4}$ sq cm per sq cm). The right sides of both curves coincide and give a maximum for the loss angle very close to experimental values both in absolute value and in its relative location. The minimum loss angle also agrees fairly closely with experimental data, although the absolute value of the loss angle, as it passes through a minimum, is somewhat higher than experimental values. The calculated curve D (Figure 2), calculated for values of cos φ, coincides with the experimental curve obtained by Clark. This is shown in Figure 4.

The coincidence of the experimental and calculated data is sufficiently close to permit the conclusion that the characteristics of the experimental curve for cos φ = f (ρ) are completely regular and can be predicted by analyzing a relatively simple equivalent circuit.

From the above-mentioned equivalent circuit it is possible to explain a series of experimental facts observed by Clark, but not explained by him in any of his articles. For example, it was observed that an increase in density and a decrease in the air penetrability of the paper results in a lowered minimum value for the loss angle. In effect, a decrease in the paper's air penetrability is evidence of the fact that the number and cross section of the pores has decreased, i.e., a lower value for S_k. Under these conditions the coefficient η increases, but this in turn causes a displacement of the curve of $\tan \delta_{\Sigma}$ = f (ρ), which assures a decrease in the value of $\tan \delta_{\Sigma}$ when it is at its minimum (see curves D and E as well as B and C in Figure 2).

Experiments showed that attempts to lower the specific resistance of the semiconducting liquids by adding water or alcohol were unsuccessful, since for a value of specific resistance of 10^{-7} ohm/cm the expected minimum for the loss angle was not observed. Quite to the contrary, a sharp increase in the loss angle occurs. It is well known that water, absorbed by the paper, increases the conductivity without forming uniform veins of water perforating the paper.

In the subject case, the improved conductivity is attributed to the fact that the walls of the pores immediately absorb the water, i.e., the cellulose itself, but the water does not completely fill the pores in the case where the paper is not directly immersed in the water. For that reason the water retained by the semiconducting liquid is selectively absorbed by the cellulose with a resulting decrease in the resistance of η R in the circuit shown in Figure 3. This is due to an increase in conductivity of the compact layer of cellulose, as well as in conductivity of the impregnating substance in the pores. These facts tend to disturb the rules obtained for the variation of $\tan \delta_x = f(\rho)$, as was observed experimentally. Alcohol, because of the presence of the hydroxyl groups, acts similarly to water.

Experiments showed that the minimum loss angle, with the same value for the specific resistance of the semiconducting substance, depends on its chemical composition, in systems not containing water or alcohol, and can fluctuate within the limits $\cos \varphi = 0.03$ to 0.10. This is explained by the fact that the penetrability of paper varies for different types of liquids, i.e., S_k and η may vary according to the type of liquid.

As mentioned above, paper condensers impregnated with semiconducting liquid may have valuable industrial applications, particularly as small-dimension low-voltage condensers for industrial frequencies. At higher voltages these condensers are unable to perform for long periods of time since the increased loss angle leads to a prohibitive overheating of the unit. Under conditions of interrupted operation, for example, for starting single-phase motors, the operating voltage can be raised as high as 400 to 500. These condensers are not suitable for DC circuits because of low insulation resistance.

Despite the fact that the field of application for impregnated paper condensers is very limited, their manufacture on an industrial scale is of technical interest since it would eventually result in smaller dimensioned paper condensers and hence lead to a decrease in the consumption of paper and foil in the production of paper condensers.

[Appended figures follow]

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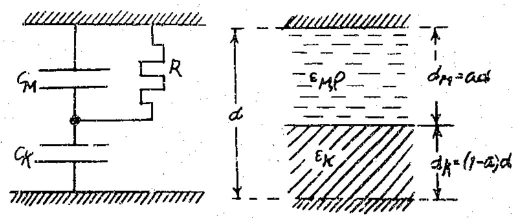

Figure 1. Equivalent Circuit Which Does not Take Into Account Presence of Open Pores in Paper

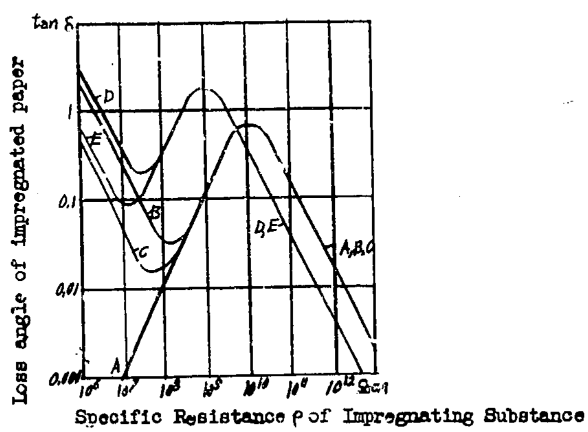

Figure 2. Curves Obtained by Calculating Relationship Between Loss Angle of Impregnated Paper and Specific Resistance of Impregnating Substance

One sheet of paper of 10-micron thickness: curve A-$\eta = 0$; curve B-$\eta = 4{,}000$; curve C-$\eta = 18{,}600$

Sheet of paper and layer of gauze of 100-micron thickness: curve D-$\eta = 100$; curve E-$\eta = 500$

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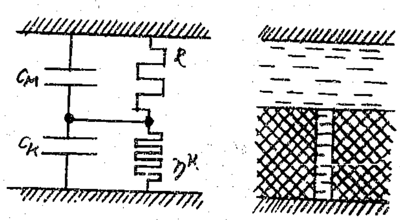
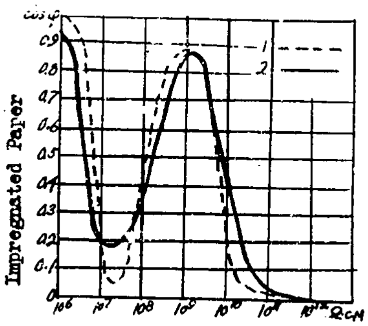


Figure 3. Equivalent Circuit, Which Takes Into Account Presence of Open Pores in Paper

Power Factor of the Impregnated Paper



Specific Resistance ρ of Impregnating Substance

Figure 4. Comparison of Experimental and Calculated Curves Showing Relationship Between cos φ of Impregnated Paper and Specific Resistance of Impregnating Substance

(1) Experimental curve; (2) calculated curve

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